

United States Patent [19]

Enomoto et al.

[54] URACIL DERIVATIVES AND THEIR HERBICIDAL USE

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[56]

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[57] ABSTRACT

A compound of the formula:



wherein \mathbb{R}^1 is a trifluoromethyl group or a pentafluoroethyl group, \mathbb{R}^2 is a methyl group or an amino group, \mathbb{R}^3 is a C₁-C₇ alkyl group, a C₃-C₇ alkenyl group, a C₃-C₇ alkynyl group, a halo (C₁-C₆) alkyl group, a halo (C₃-C₆) alkenyl group or a C₁-C₄ alkoxy(C₁-C₃)alkyl group and Y is an oxygen atom or a sulfur atom, which is useful as a herbicide.

11 Claims, No Drawings

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URACIL DERIVATIVES AND THEIR HERBICIDAL USE

This application is a divisional of copending applica- 5 tion Ser. No. 07/585,917, filed on Sep. 21, 1990, now abandoned, the entire contents of which are hereby incorporated by reference.

The present invention relates to uracil derivatives, and their production and use. More particularly, it re- 10 lates to uracil derivatives, a process for producing them, and their use as herbicides.

EP-A-311135 discloses some uracil derivatives useful as herbicides. However, these known herbicides are not sufficient in herbicidal potency or have poor selectivity 15 between crop plants and weeds. Their herbicidal activity is thus not necessarily satisfactory.

It has now been found that uracil derivatives of the formula:



wherein \mathbb{R}^1 is a trifluoromethyl group or a pentafluoroethyl group, R² is a methyl group or an amino group, \mathbb{R}^3 is a C₁-C₇ alkyl group, a C₃-C₇ alkenyl group, a C₃-C₇ alkynyl group, a halo(C₁-C₆)alkyl group, a ha $lo(C_3-C_6)$ alkenyl group or a C₁-C₄ alkoxy(C₁-C₃)alkyl group and Y is an oxygen atom or a sulfur atom, show ³⁵ a high herbicidal potency against various weeds with a high selectivity between crop plants and weeds. Thus, they produce a strong herbicidal activity against a wide variety of weeds including broad-leaved weeds, Graminaceous weeds, Commelinaceous weeds and 40 Cyperaceous weeds in agricultural plowed fields by foliar or soil treatment without producing any material phytotoxicity on various agricultural crops such as corn, wheat, barley, rice plant, soybean and cotton. Examples of the broad-leaved weeds include wild buck- 45 wheat (Polygonurn convolvulus), pale smartweed (Polygonum lapathifolium), common pursiane (Portulaca oleracea), common chickweed (Stellaria media), common lambsquarters (Chenopodium album), redroot pigweed (Amaranthus retroflexus), radish (Raphanus sativus), 50 wild mustard (Sinapis arvensis), shepherdspurse (Capsella bursapastoris), hemp sesbania (Sesbania exaltata), sicklepod (Cassia obtusifolia), velvetleaf (Abutilon theophrasti), prickly sida (Sida soinosa), field pansy (Viola arvensis), catchweed bedstraw (Galium aparine), ivyleaf 55 mornigglory (Ipomoea hederacea), tall morningglory (Ipomoea purpurea), field bindweed (Convolvulus arvensis), henbit (Lamium amplexicaure), jimsonweed (Datura stramonium), black nightshade (Solanum nigrum), persian speedwell (Veronica persica), common cockle- 60 drohalogenating agent are used respectively in amounts bur (Xanthium pensylvanicum), common sunflower (Helianthus annuus), scentless chamomile (Matricaria perforata), corn marigold (Chrysanthemum segetum), purple deadnettle (Lamium purpureum), sun spurge (Euphorbia

Examples of Graminaceous weeds include Japanese millet (Echinochloa frumentacea), barnyardgrass (Echinochloa crus-galli), green foxtail (Setaria viridis),

large crabgrass (Digitaria sanguinalis), annual bluegrass (Poa annua), blackgrass (Alopecurus myosuroides), oats (Avena sativa), wild oats (Avena fatua), johnsongrass (Sorghum halepense), quackgrass (Agropyron repens), bermudagrass (Cynodon dactylon), downy brome (Bromus tectorum), giant foxtail (Setaria faberi), etc. Example of Commelinaceous weeds include asiatic dayflower (Commelina communis), etc. Examples of Cyperaceous weeds include rice flatsedge (Cyperus iria), purple nutsedge (Cyperus rotundus), etc.

The uracil derivatives (I) of the invention are also effective in exterminating paddy field weeds including Graminaceous weeds such as barnyardgrass (Echinochloa oryzicola), broad-leaved weeds such as common falsepimpernel (Lindernia procumbens), indian toothcup (Rotala indica) and waterwort (Elatine triandra), Cyperaceous weeds such as water nutgrass (Cyperus serotinus), hardstem bulrush (Scirupus juncoides), needle spikerush

20 (Eleocharis acicularis) and umbrella sedge (Cyperus difformis), and others such as monochoria (Monochoria vaginalis) and arrowhead (Sagittaria pygmaea) without producing any phytotoxicity to rice plants on flooding treatment.

Among the compounds (I), preferred are those wherein \mathbb{R}^1 , \mathbb{R}^2 and \mathbb{R}^3 are each as defined above and Y is a sulfur atom. More preferred are those wherein \mathbb{R}^1 and R^2 are each as defined above, R^3 is a C₁-C₇ alkyl group and Y is a sulfur atom.

Typical examples of the preferred compounds are 1-[6-fluoro-3-sec-butyl-2 (3H) -benzothiazolon-5-yl]-3methyl-4-trifluoromethyl -1,2,3,6-tetrahydropyrimidine-2,6-dione, 1- [6-fluoro-3-isopropyl-2 (3H) -benzothiazolon-5-yl]-3-methyl-4-trifluoromethyl-1,2,3,6-tetrahydropyrimidine-2,6-dione, etc.

The compound (I) is obtainable by the procedures as set forth below.

Procedure (A):

The compound (R^2 —CH₃) is obtainable by reacting a compound of the formula:

(II)



wherein R¹ R³ and Y are each as defined above with a methylating agent.

The reaction is usually carried out in the presence of a dehydrohalogenating agent in an inert solvent at a temperature of about 20° to 100° C. for a period of about 0.5 to 8 hours.

In general, the methylating agent and the dehyof about to 1.2 equivalents and of about 1 to 3 equivalents to one equivalent of the compound (II). As the methylating agent, there may be used methyl iodide, methyl bromide, methyl chloride, dimethyl sulfate or helioscopia), spotted spurge (Euphorbia maculata), etc. 65 the like. Examples of the dehydrohalogenating agent are organic bases (e.g. pyridine, triethylamine, N,N-diethylaniline), inorganic bases (e.g. sodium hydroxide, potassium hydroxide, sodium carbonate, potassium car-

bonate, sodium hydride), alkali metal alkoxides (e.g. sodium methoxide, sodium ethoxide), etc.

Examples of the inert solvent are aliphatic hydrocarbons (e.g. hexane, heptane, ligroin, petroleum ether), 5 aromatic hydrocarbons (e.g. benzene, toluene, xylene), halogenated hydrocarbons (e.g. chloroform, carbon tetrachloride, dichloroethane, chlorobenzene, dichlorobenzene), ethers (e.g. diethyl ether, diisopropyt ether, dioxane, tetrahydrofuran, diethylene glycol dimethyl 10 ether), ketones (e.g. acetone, methyl ethyl ketone, methyl isobutyl ketone, isophorone, cyclohexanone), tertiary amines (e.g. pyridine, triethylamine, N,N-diethylaniline, tributylamine, N-methylmorpholine), acid amides (e.g. formamide, N,N-dimethylformamide, acet- 15 amide), sulfur compounds (e.g. dimethytsulfoxide, sulphorane), water, etc. These may be used solely or in combination.

After completion of the reaction, the reaction mixture is subjected to ordinary post-treatment. For instance, the reaction mixture is poured into water, and the precipitated crystal are collected by filtration. Alternatively, the reaction mixture is shaken in combination with water and a water-immiscible organic solvent 25 for extraction, and the extract is concentrated. If desired, any conventional purification procedure such as chromatography, distillation or recrystallization may be applied to the resulting product.

Procedure (B):

The compound $(R^2 = NH_2)$ is obtainable by reacting the compound (II) with an aminating agent.

The reaction is usually carried out in an inert solvent at a temperature of about 20° to 100° C. for a period of $_{35}$ about 0.5 to 8 hours.

The aminating agent is normally used in an amount of about 1 to 3 equivalents to one equivalent of the compound (II). As the aminating agent, there may be used 2,4-dinitrophenoxyamine, etc.

Examples of the inert solvent are aliphatic hydrocarbons (e.g. hexane, heptane, ligroin, petroleum ether), aromatic hydrocarbons (e.g. benzene, toluene, xylene), halogenated hydrocarbons (e.g. chloroform, carbon tetrachloride, dichloroethane, chlorobenzene, dichlorobenzene), ethers (e.g. diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran, diethylene glycol dimethyl ether), ketones (e.g. acetone, methyl ethyl ketone, methyl isobutyl ketone, isophorone, cyclohexanone), 50 tertiary amines (e.g. pyridine, triethylamine, N,N-diethylaniline, tributylamine, N-methylmorpholine), acid amides (e.g. formamide, N,N-dimethylformamide, acetamide), sulfur compounds (e.g. dimethylsulfoxide, sulphorane), water, etc. 55

After completion of the reaction, the reaction mixture is subjected to ordinary post-treatment. For instance, the reaction mixture is poured into water, and the precipitated crystals are collected by filtration. Alternatively, the reaction mixture is shaken in combination with water and a water-immiscible organic solvent for extraction, and the extract is concentrated. If desired, any conventional purification procedure such as chromatography, distillation or recrystallization may be 65 applied to the resulting product.

According to the above procedure (A) or (B), the compounds (I) as shown in Table 1 are obtained.

	TABLE 1						
		$ \begin{array}{c} F & O \\ N \\ N \\ N \\ N \\ N \\ R^2 \end{array} $	(1)				
R ¹	R ²	R ³	Y				
CF3 CF3 CF3 CF3 CF3 CF3 CF3 CF3 CF3 CF3	CH ₃ CH ₃	CH ₃ C ₂ H ₅ (n)C ₃ H ₇ (i)C ₃ H ₇ (i)C ₄ H ₉ (s)C ₄ H ₉ (i)C ₄ H ₉ (i)C ₄ H ₉ (n)C ₅ H ₁₁ (n)C ₅ H ₁₃ (n)C ₇ H ₁₅ CH ₂ CH=CH ₂ CH ₂ CH=CH ₂	S S S S S S S S S S S S S S S S				
CF3	CH3	CH ₂ C=CH H CH ₃	S				
CF3	CH3	CH(CH ₃)C=CH H CH ₃	S				
CF3	CH3	CH ₂ C=CCH ₃ H H	S				
CF ₃	CH3	CH(CH ₃)C=CCH ₃ H H	S				
CF_3 CF_3	$\begin{array}{c} {\rm CH}_3 \\ {\rm C$	$\begin{array}{l} CH_2CH=C(CH_3)_2\\ CH(CH_3)CH=C(CH_3)_2\\ CH=C=CH_2\\ CH_2C\equiv CH\\ CH(CH_3)C\equiv CH\\ CH_2C\equiv CC_1\\ GH_2C\equiv CC_2\\ GH_3C\equiv CC_2\\ H_3\\ CH_2C\equiv CC_2\\ H_5\\ CH_2C\equiv CC_2\\ H_5\\ CH_2C\equiv CC_3\\ H_7(n)\\ CH_2C=C_3\\ H_7(n)\\ CH_2C=C_3\\ H_7(n)\\ CH_2CC_4\\ H_9(n)\\ CH_2C$	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~				
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		5					6	
	Т	ABLE 1-continued				Т	ABLE 1-continued	
		FO		(I)			FO	(I)
	у —{	$-R^1$		5		ч — 🦑	$\sim N = R^1$	
	_L `	∖∕ ∖N′				Ľ,	∑N	
	<u>را / ا</u>					<u>م</u> ار م		
	- N	- K*				n N	- K -	
	R ²	3		10		R		
R ¹	R ²	R ³	Y		R ¹	R ²	R ³	Y
CF ₃	CH ₃	СH2C=СН	s		C_2F_5	CH ₃	CH ₂ C=CCl	S
		I I H Cl		15			I I H H	
	~	CII	-	15	CaEc	CH		s
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₃	CH3 C2H5	S		021 5	City	$CH_2C=CH$	5
C_2F_5	CH ₃	(n)C ₃ H ₇	S				Ĥ Ċl	
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₃	(1)C3H7 (n)C4H9	S	20	CF ₃	NH ₂	CH ₃	s
C_2F_5	CH ₃	(s)C4H9	S	20	CF3	NH ₂	C_2H_5	S
C ₂ F ₅	CH ₃	(i)C ₄ H ₉ (n)C ₆ H ₁₁	S		CF ₃	NH ₂	(i)C ₃ H ₇	S
C_2F_5	CH ₃	$(n)C_{6}H_{13}$	s		CF ₃	NH ₂	(n)C4H9	S
C_2F_5	CH ₃	(n)C7H15	s		CF3 CF2	NH ₂ NH ₂	(s)C4H9 (i)C4H9	S
C ₂ F ₅	CH ₃	CH ₂ CH=CH ₂	S	25	CF ₃	NH ₂	$(n)C_5H_{11}$	ŝ
0215	City		0		CF3	NH ₂	$(n)C_6H_{13}$	S
C_2F_5	CH ₃	CH ₂ C=CH	S		CF ₃	NH ₂	$CH_2CH=CH_2$	S
					CF ₃	NH_2	CH(CH ₃)CH=CH ₂	S
				30	CF ₃	NH_2	сн.с=сн	s
C_2F_5	CH ₃	CH(CH ₃)C=CH	S					
		H CH3					н СН3	
	~~~		-		$CF_3$	$\mathbf{NH}_2$	CH(CH ₃ )C=CH	S
$C_2F_5$	CH ₃	$CH_2C = CCH_3$	5	35			 H CH3	
		H H					,	
CaEs	CH ₂		s		CF ₃	$NH_2$	CH ₂ C=CCH ₃	S
-2- 5	0115	$CH(CH_3)C=CCH_3$	-				I I H H	
		нн		40	CF ₂	NHa		s
C ₂ F ₅	CH ₃	CH ₂ CH=C(CH ₃ ) ₂	S		0. 3	1.112	$CH(CH_3)C=CCH_3$	ũ
C ₂ F ₅	CH ₃	$CH(CH_3)CH=C(CH_3)_2$	S				Ĥ Ĥ	
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₂	$CH = C = CH_2$ $CH_2C = CH$	s s		CF ₃	NH ₂	CH ₂ CH=C(CH ₂ ) ₂	s
$C_2F_5$	CH ₃	CH(CH ₃ )C=CH	s	45	CF ₃	NH ₂	$CH(CH_3)CH=C(CH_3)_2$	s
C ₂ F ₅	CH ₃	CH ₂ C CCH ₃	Ś	45	CF ₃	NH ₂	CH=C=CH ₂	S
C ₂ F ₅	CH ₃	$CH_{CH_3}C \equiv CC_{H_3}$	S		CF3 CF3	NH ₂ NH ₂		s
$C_2F_5$	CH ₃	$CH_2CE_2CC_2H_5$ $CH_3CE_2CC_2H_5$	ŝ		CF ₃	NH ₂	CH ₂ C CCH ₃	s
$C_2F_5$	CH ₃	$CH_2C \equiv CC_3H_7(n)$	S		CF ₃	NH ₂	CH(CH ₃ )C=CCH ₃	S
$C_2F_5$	CH ₃	CH(CH ₃ )C=CC ₃ H ₇ (n)	S	50	CF3 CF3	NH ₂ NH ₂	$CH_2C = CC_2H_5$ $CH(CH_3)C = CC_2H_5$	s
C2F5 C2F5	CH3 CH3	CH ₂ OCH ₃ CH ₂ OC ₂ H ₅	s		CF ₃	NH ₂	$CH_2C \equiv CC_3H_7(n)$	s
$C_2F_5$	CH ₃	$CH_2OC_3H_7(n)$	S		CF3	NH ₂	CH(CH ₃ )C=CC ₃ H ₇	S
C ₂ F ₅	CH ₃	CH ₂ OC ₃ H ₇ (i)	S		CF3 CF3	NH ₂ NH ₂	CH ₂ OC ₂ H ₅	S
	CH ₃ CH ₂	$CH_2OC_4H_9(n)$ $CH_2OC_4H_9(s)$	S		CF ₃	NH ₂	CH ₂ OC ₃ H ₇ (n)	S
$C_2F_5$	CH ₃	$CH_2OC_4H_9(i)$	Š	55	CF3	NH ₂	$CH_2OC_3H_7(i)$	S
$C_2F_5$	CH ₃	CH(CH ₃ )OC ₂ H ₅	S		CF3	NH ₂	CH ₂ OC ₄ H ₉ (II) CH ₂ OC ₄ H ₉ (s)	S
C ₂ F ₅	CH ₃	$CH(CH_3)OC_3H_7(n)$	S		$CF_3$	NH ₂	CH ₂ OC ₄ H ₉ (i)	S
C2F5 C2F5	CH3 CH3	CH(CH ₃ )OC ₄ H ₉ (n)	s		CF3	NH ₂	CH(CH ₃ )OCH ₃	S
$C_2F_5$	CH ₃	CH(CH ₃ )OC ₄ H ₉ (s)	S	60	CF3	NH ₂	CH(CH ₃ )OC ₃ H ₇ (n)	S
C ₂ F ₅	CH ₃	CH(CH ₃ )OC ₄ H ₉ (i)	S	- •	CF ₃	NH ₂	CH(CH ₃ )OC ₃ H ₇ (i)	S
C2F5 C2F5	CH3 CH2	C2H4OCH3 C2H4OC2H5	5 5		CF3	NH2	CH(CH ₃ )OC ₄ H ₉ (n)	S
$C_2F_5$	CH ₃	CH(CH ₃ )CH ₂ OC ₂ H ₅	ŝ		CF ₃	NH ₂	CH(CH ₃ )OC ₄ H ₉ (i)	S
$C_2F_5$	CH ₃	CH ₂ CH ₂ F	S		$CF_3$	NH ₂	C ₂ H ₄ OCH ₃	S
C2F5 C2F5	CH3 CH1	CH2CF3 CH(CH2)CH2F	5 5	65	CF ₃ CF ₂	NH2 NH2	C2H4OC2H5 CH(CH2)CH2OC2He	S
$C_2F_5$	CH ₃	CH(CH ₃ )CF ₃	ŝ		CF ₃	NH ₂	CH ₂ CH ₂ F	ŝ
$C_2F_5$	CH ₃	$CH_2CCl=CH_2$	s		CF ₃	NH ₂	CH ₂ CF ₃	S
					CF3	INF12	Cn(Cn3)Ch2r	3

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	1	E O		m	· · · · · · · · · · · · · · · · · · ·	1	F O	(1)
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	o N	$O R^2$				o _N	0 _R 2	
	I R	3		10		l R ²	3	
R ¹	R ²	R ³	Y		R1	R ²	R ³	Y
CF ₃	NH2 NH2	CH(CH ₃ )CF ₃ CH ₂ CCI=CH ₂	S		C ₂ F ₅	NH2 NH2	CH ₂ CH ₂ F	S
Cr3	i i i i z		5	15	$C_2F_5$ $C_2F_5$	NH ₂	CH(CH ₃ )CH ₂ F	S
CF ₃	$NH_2$	CH ₂ C=CCl	s	15	C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	$CH(CH_3)CF_3$ $CH_2CCI=CH_2$	S S
		H H			-2- 5			-
CE ₂	NHa		s		$C_2F_5$	NH ₂	CH ₂ C=CCI	S
Cr y	1112	$CH_2C = CH$	0	20			H H	
		H Cl		20	C ₂ F ₅	NH2		S
$C_2F_5$	NH ₂	CH ₃	S		25	2		-
C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	C ₂ H ₅ (n)C ₃ H ₇	S S				H Cl	
$C_2F_5$	NH ₂	(i)C ₃ H ₇	S	25	CF ₃	CH3	CH ₃	0
$C_2F_5$ $C_2F_5$	$NH_2$ $NH_2$	(n)C4H9 (s)C4H9	S	40	CF3 CF3	CH ₃ CH ₃	$C_2H_5$ (n)C_3H_7	0
$C_2F_5$	NH ₂	(i)C4H9	S		CF ₃	CH ₃	(i)C ₃ H ₇	0
$C_2F_5$ $C_2F_5$	$\mathbf{NH}_2$ $\mathbf{NH}_2$	$(n)C_5H_{11}$ $(n)C_6H_{13}$	S		CF3 CF3	CH3 CH3	(n)C4H9 (s)C4H9	0
$C_2F_5$	NH ₂	(n)C7H15	S	20	CF ₃	CH ₃	(i)C4H9	Ō
C ₂ F ₅ C ₂ F ₅	NH2 NH2	$CH_2CH=CH_2$ $CH(CH_3)CH=CH_2$	s s	50	CF ₃ CF ₃	CH3 CH3	$(n)C_{5}H_{11}$ $(n)C_{6}H_{13}$	0
-2- 5		(5)2	-		CF ₃	CH ₃	$(n)C_7H_{15}$	Ō
$C_2F_5$	NH ₂	$CH_2C = CH$	S		CF3 CF3	CH3 CH3	$CH_2CH=CH_2$ $CH(CH_3)CH=CH_2$	0
		H CH ₃			J			C .
C.F.	NTLL.		0	35	CF ₃	CH3	CH ₂ C=CH	0
C21'5	14112	$CH(CH_3)C = CH$	3				H CH ₃	
		Ĥ ĊH₃			CE	CH		0
C ₂ F ₅	NH ₂	CH-C-CCH-	S		Cr 3	CII3	$CH(CH_3)C = CH$	0
2.0	~			40			Ĥ CH ₃	
		НН			CF ₃	CH ₃	CH-C-CCH-	0
$C_2F_5$	$NH_2$	CH(CH ₃ )C=CCH ₃	S		-	Ū.		
							н н	
		11 11		45	CF ₃	$CH_3$	$CH(CH_3)C = CCH_3$	0
C ₂ F ₅ C ₂ F ₅	NH2 NH2	$CH_2CH=C(CH_3)_2$ $CH(CH_3)CH=C(CH_3)_3$	S					
$C_2F_5$	NH ₂	CH=C=CH ₂	ŝ					
C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	$CH_2C \equiv CH$ $CH(CH_2)C = CH$	S		CF ₃ CF ₂	CH ₃ CH ₂	$CH_2CH = C(CH_3)_2$ $CH(CH_2)CH = C(CH_2)_2$	0
$C_2F_5$	NH ₂	CH ₂ C=CCH ₃	S	50	CF ₃	CH ₃	CH=C=CH ₂	Ö
C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	$CH(CH_3)C \equiv CCH_3$ $CH_2C \equiv CC_2H_4$	S S		CF ₃ CF ₂	CH ₃ CH ₂	CH ₂ C=CH CH(CH ₂ )C=CH	0
$C_2F_5$	NH ₂	$CH(CH_3)C \equiv CC_2H_5$	ŝ		CF ₃	CH ₃	CH ₂ C≡CCH ₃	ŏ
C ₂ F ₅	NH ₂ NH ₂	$CH_2C \equiv CC_3H_7(n)$ $CH(CH_2)C \equiv CC_2H_7(n)$	S		CF ₃ CF ₂	CH ₃ CH ₂	$CH(CH_3)C \equiv CCH_3$ $CH_2C \equiv CC_2H_4$	0
$C_2F_5$	NH ₂	CH ₂ OCH ₃	s	55	CF ₃	CH ₃	CH(CH ₃ )C=CC ₂ H ₅	ŏ
C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	$CH_2OC_2H_5$ $CH_2OC_2H_7(n)$	S		CF3 CF7	CH ₃ CH ₂	$CH_2C \equiv CC_3H_7(n)$ $CH(CH_2)C \equiv CC_2H_7(n)$	0
$C_2F_5$	NH ₂	$CH_2OC_3H_7(i)$	Š		CF ₃	CH ₃	CH ₂ OCH ₃	õ
C ₂ F ₅ C ₂ F ₅	NH ₂ NH ₂	$CH_2OC_4H_9(n)$ $CH_2OC_4H_9(s)$	S S		CF ₃ CF ₂	CH3 CH2	$CH_2OC_2H_5$ $CH_2OC_2H_7(n)$	0
$C_2F_5$	NH ₂	CH ₂ OC ₄ H ₉ (i)	Š	60	CF ₃	CH ₃	CH ₂ OC ₃ H ₇ (i)	Ō
C ₂ F ₅	NH2 NH2	CH(CH ₃ )OCH ₃ CH(CH ₂ )OC ₂ H ₅	S		CF ₃ CF ₂	CH ₃ CH ₂	CH ₂ OC ₄ H ₉ (n) CH ₂ OC ₄ H ₉ (s)	0
$C_2F_5$	NH ₂	CH(CH ₃ )OC ₃ H ₇ (n)	ŝ		CF ₃	CH ₃	CH2OC4H9(i)	ŏ
C ₂ F ₅	NH ₂ NH ₂	CH(CH ₃ )OC ₃ H ₇ (i) CH(CH ₂ )OC ₄ H ₂ (n)	S		CF ₃	CH3	CH(CH ₃ )OCH ₃ CH(CH ₃ )OC ₂ H ₂	0
$C_2F_5$	NH ₂	CH(CH ₃ )OC ₄ H ₉ (s)	S	65	CF ₃	CH ₃	CH(CH ₃ )OC ₃ H ₇ (n)	õ
C ₂ F ₅	NH2	CH(CH ₃ )OC ₄ H ₉ (i) C ₂ H ₄ OCH ₂	S		CF3	CH3	CH(CH ₃ )OC ₃ H ₇ (i)	0
$C_2 F_5$ $C_2 F_5$	NH ₂	$C_2H_4OC_2H_5$	S		CF ₃	CH ₃	CH(CH ₃ )OC ₄ H ₉ (s)	õ
$C_2F_5$	NH ₂	CH(CH ₃ )CH ₂ OC ₂ H ₅	S		CF ₃	CH ₃	CH(CH3)OC4H9(i)	0

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R ¹	R ²	R ³	Y		R ¹	R ²	R ³	Y
CF ₃ CF ₃	CH3 CH3	C ₂ H ₄ OCH ₃ C ₂ H ₄ OC ₂ H ₅	0		C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₃	$CH(CH_3)OC_4H_9(n)$ $CH(CH_3)OC_4H_9(s)$	0
CF ₃	CH ₃	CH(CH ₃ )CH ₂ OC ₂ H ₅	0	15	C ₂ F ₅	CH ₃	CH(CH ₃ )OC ₄ H ₉ (i)	0
CF ₃	CH ₃	CH ₂ CF ₃	ŏ		$C_2F_5$	CH ₃	$C_2H_4OC_2H_5$	0
CF3 CF3	CH3 CH3	CH(CH ₃ )CH ₂ F CH(CH ₃ )CF ₃	0		$C_2F_5$ $C_2F_5$	CH3 CH3	CH(CH ₃ )CH ₂ OC ₂ H ₅ CH ₂ CH ₂ F	0 0
CF ₃	CH ₃	CH ₂ CCl=CH ₂	0		C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₂	CH ₂ CF ₃ CH(CH ₂ )CH ₂ E	0
CF ₃	CH ₃	CH2C=CCl	0	20	$C_2F_5$	CH ₃	CH(CH ₃ )CF ₃	ŏ
		 H H			$C_2F_5$	CH3	CH ₂ CCI=CH ₂	U
CE	CH		0		$C_2F_5$	$CH_3$	CH ₂ C=CCl	0
C1 3	C113	$CH_2C = CH$	0	25			H H	
		H Cl		25	$C_2F_5$	CH ₃	CH ₂ C=CH	о
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₂	CH3 CaHs	0					
$C_2F_5$	CH ₃	(n)C ₃ H ₇	ŏ		<b>CT</b>			<u> </u>
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₃	(1)C3H7 (n)C4H9	0	30	CF ₃ CF ₃	NH2 NH2	CH3 C2H5	0
C2F5 C2F5	CH ₃ CH ₂	(s)C4H9 (i)C4H9	0		CF ₃ CF ₃	NH ₂ NH ₂	(n)C3H7 (i)C3H7	0
C ₂ F ₅	CH ₃	$(n)C_5H_{11}$	ŏ		CF ₃	NH ₂	(n)C4H9	Ö
$C_2F_5$ $C_2F_5$	CH ₃ CH ₃	$(n)C_7H_{15}$	ő		CF3 CF3	NH ₂ NH ₂	(i)C4H9	0
C ₂ F ₅ C ₂ F ₅	CH3 CH3	$CH_2CH=CH_2$ $CH(CH_3)CH=CH_2$	0	35	CF ₃ CF ₃	NH ₂ NH ₂	$(n)C_5H_{11}$ $(n)C_6H_{13}$	0
CaFe	CH		0		CF ₃	NH ₂	$(n)C_7H_{15}$	0
0215	Chiş	$CH_2C = CH$	0		CF ₃	NH ₂	$CH_2CH_2CH_2$ CH(CH_3)CH=CH_2	ŏ
		H CH ₃		40	CF ₃	NH ₂	CH-C=CH	ο
$C_2F_5$	CH ₃	СН(СН3)С=СН	0					
		I I H CH3					ii cii,	
CaFe	CH		0		CF ₃	NH ₂	CH(CH ₃ )C=CH	0
C21.2	C113	$CH_2C=CCH_3$	U U	45			н снз	
		нн			CF ₃	NH ₂	CH ₂ C=CCH ₂	о
$C_2F_5$	CH ₃	CH(CH ₃ )C=CCH ₃	0					
		H H		50	CE	****		0
C ₂ F ₅	CH ₃	CH ₂ CH=C(CH ₃ ) ₂	0	50	CF3	NH ₂	$CH(CH_3)C = CCH_3$	0
C ₂ F ₅	CH ₃ CH ₂	$CH(CH_3)CH=C(CH_3)_2$ $CH=C=CH_2$	0				н н	
$C_2F_5$	CH ₃	CH ₂ C≡CH	0		CF ₃	NH ₂	CH ₂ CH=C(CH ₃ ) ₂	0
C ₂ F ₅ C ₂ F ₅	CH ₃ CH ₃	$CH(CH_3)C = CH$ $CH_2C = CCH_3$	0	55	CF ₃ CF ₃	NH2 NH2	$CH(CH_3)CH=C(CH_3)_2$ $CH=C=CH_2$	0
C2F5 C2F6	CH ₃ CH ₂	$CH(CH_3)C \equiv CCH_3$ $CH_2C \equiv CC_2H_5$	0		CF ₃ CF ₂	NH2 NH2	$CH_2C \equiv CH$ $CH(CH_2)C \equiv CH$	0
$C_2F_5$	CH ₃	CH(CH ₃ )C≡CC ₂ H ₅	ŏ		CF ₃	NH ₂	CH ₂ C≡CCH ₃	Õ
C2F5 C2F5	CH ₃ CH ₃	$CH_2C = CC_3H_7(n)$ CH(CH_3)C = CC_3H_7(n)	0		CF ₃ CF ₃	NH2 NH2	$CH(CH_3)C \equiv CCH_3$ $CH_2C \equiv CC_2H_5$	0
C ₂ F ₅ C ₂ F ₅	CH3 CH3	CH2OCH3 CH2OC2H5	0	60	CF3 CF3	NH2 NH2	$CH(CH_3)C \equiv CC_2H_5$ $CH_2C \equiv CC_3H_7(n)$	0 0
C ₂ F ₅	CH ₃	$CH_2OC_3H_7(n)$	õ		CF3	NH ₂	CH(CH ₃ )C=CC ₃ H ₇ (n)	Ō
$C_2F_5$ $C_2F_5$	CH ₃	CH ₂ OC ₄ H ₉ (n)	o		CF3 CF3	NH ₂	CH ₂ OC ₂ H ₅	0
C2F5 C2F5	CH3 CH3	CH2OC4H9(s) CH2OC4H9(i)	0 0	65	CF3 CF3	NH2 NH2	$CH_2OC_3H_7(n)$ $CH_2OC_3H_7(i)$	0 0
C ₂ F ₅	CH ₃	CH(CH ₃ )OCH ₃	0	05	CF3	NH ₂	CH2OC4H9(n)	0
$C_2F_5$ $C_2F_5$	CH ₃	$CH(CH_3)OC_3H_7(n)$	0		CF3 CF3	NH ₂	$CH_2OC_4H_9(s)$ $CH_2OC_4H_9(i)$	0
C ₂ F ₅	$CH_3$	CH(CH ₃ )OC ₃ H ₇ (i)	0		$CF_3$	$NH_2$	CH(CH ₃ )OCH ₃	0

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			$\rightarrow$ $\gamma$ $\gamma$ $N$ $N$ $\gamma$ $N$ $N$ $\gamma$ $N$					$\rightarrow$ $\rightarrow$ $N$ $\downarrow$ $N$	
		R ³	K		10		I R ³	ĸ	
	$\mathbb{R}^1$	R ²	R ³	Y		$\mathbb{R}^1$	R ²	R ³	
_	CF ₃	NH ₂	CH(CH ₃ )OC ₂ H ₅	0		C ₂ F ₅	NH ₂	CH2OC4H9(s)	-
	CF ₃	NH ₂	CH(CH ₃ )OC ₃ H ₇ (n)	0		$C_2F_5$	NH ₂	CH ₂ OC ₄ H ₉ (i)	
	CF3 CF3	NH2 NH2	$CH(CH_3)OC_3H_7(1)$ $CH(CH_3)OC_4H_9(n)$	0	15	C ₂ F ₅	NH2 NH2	CH(CH3)UCH3	
	CF ₃	NH ₂	CH(CH3)OC4H9(II)	ŏ		$C_2F_5$	NH ₂	CH(CH ₂ )OC ₂ H ₇ (n)	
	CF ₃	NH ₂	CH(CH ₃ )OC ₄ H ₉ (i)	ō		$C_2F_5$	NH ₂	CH(CH ₃ )OC ₃ H ₇ (i)	
	$CF_3$	NH ₂	C ₂ H ₄ OCH ₃	0		$C_2F_5$	NH ₂	CH(CH ₃ )OC ₄ H ₉ (n)	
	CF ₃	$NH_2$	C ₂ H ₄ OC ₂ H ₅	0		$C_2F_5$	$NH_2$	CH(CH3)OC4H9(s)	
	CF3	NH ₂	CH(CH ₃ )CH ₂ OC ₂ H ₅	0	20	$C_2F_5$	NH ₂	CH(CH ₃ )OC ₄ H ₉ (i)	
	CF3	NH2 NH2	CH ₂ CH ₂ F	0	20	C ₂ F ₅	NH ₂	C ₂ H ₄ OC ₁ H ₂	
	CF ₃	NH ₂	CH(CH ₂ )CH ₂ F	0		$C_2 \Gamma_5$	NH ₂	CH(CH ₂ )CH ₂ OC ₂ H	íc
	CF ₃	NH ₂	CH(CH ₃ )CF ₃	ŏ		C2F5	NH ₂	CH ₂ CH ₂ F	Ċ.
	$CF_3$	$\mathbf{NH}_{2}$	CH2CCI=CH2	0		$C_2F_5$	NH ₂	$CH_2CF_3$	
						$C_2F_5$	NH ₂	CH(CH ₃ )CH ₂ F	
	$CF_3$	NH ₂	CH ₂ C=CCl	0	25	$C_2F_5$	NH ₂	CH(CH ₃ )CF ₃	
						$C_2 F_5$	NH ₂	$CH_2CCI = CH_2$	
			n n			C ₂ F ₅	NH ₂		
	CF ₃	NH ₂	CHACTCH	о		-2- 5	2	$CH_2C = CCI$	
								н н	
			Ĥ Ĉl		30	0.5			
	C.F.	NILL.	CH	0		$C_2F_5$	NH ₂	$CH_2C=CH$	
	C ₂ F ₅	NH ₂	CoHs	ŏ					
	$C_2F_5$	NH ₂	(n)C ₃ H ₇	ŏ			••••••		
	$C_2F_5$	NH ₂	(i)C ₃ H ₇	0		_			
	$C_2F_5$	NH ₂	(n)C4H9	0	35	Some of	of the cor	npounds (I) have	;
	$C_2F_5$	NH ₂	(s)C4H9	0		which are	e also inclu	uded within the so	:0
	C ₂ F ₅	NH2 NH2	$(1)C_4H_9$	0		tion.			
	$C_2F_5$	NH ₂	(n)C6H13	ŏ		Typical	l embodin	nents for product	tic
	$C_2F_5$	NH ₂	$(n)C_7H_{15}$	ō		pounds (]	I) are illu	stratively shown	iı
	$C_2F_5$	NH ₂	CH ₂ CH=CH ₂	0	40	Examples		•	
	$C_2F_5$	$NH_2$	$CH(CH_3)CH=CH_2$	0		Exampl	le 1		
	CaFe	NH ₂		0		To a so	lution of 1	I-J3-sec-butyl-6-flu	ю
	024 3	11112	$CH_2C=CH$	0		thiazolon-	-5-yl]-4-tri	fluoromethyl-1,2,3	i,6
			h CH3			dropyrimi	idine-2.6-d	ione (2.0 g) in di	m
					45	(10 g), m	ethvl iodic	(0.5 g) and pot	a
	$C_2F_5$	$NH_2$	CH(CH ₃ )C=CH	0		(1.6 g) w	vere adde	d, and the result	a
						heated at	40° to 80°	C. for 3 hours. Af	Ìŧ
			н Сн3			the reaction	on, the rea	action mixture was	s ·
	CaFe	NHa		0		ter, and	the precir	itated crystals w	'e
	021 3	11112	$CH_2C = CCH_3$	Ũ	50	filtraction	. washed a	and dried. The res	ic
			н н			by colum	n chroma	tography to give	
						fluoro-2(3	H) -ber	zothiazolon-5-vl]	-3
	$C_2F_5$	$NH_2$	$CH(CH_3)C=CCH_3$	0		fluoromet	hvl-1.2.3.6	tetrahvdropyrim	iđ
						(1.0 g).		······································	
			пп		55	H-NMI	Rδ(ppm)	[CDCl3, 60 MHz]	: (
	$C_2F_5$	NH ₂	$CH_2CH=C(CH_3)_2$	О		3H), 1.48	(d, J = 7 H)	$z_{1}$ 3H) 1.6–2.3 (m.	?
	$C_2F_5$	$NH_2$	$CH(CH_3)CH=C(CH_3)_2$	0		4.0-4.5 (m	. 1H). 6.3	(s. 1H), 7.03 (d. J	_
	$C_2F_5$	NH ₂	$CH=C=CH_2$	0		(d. $J = 10$	Hz. 1H).	(0,), (2, 0	
	C2F5	NH2 NH2		U A		Exampl	e 2		
	$C_2F_5$	NH ₂	CH ₂ C=CH ₂	õ	60	Toas	olution of	1-[6-fluoro-3-pro	'n
	$C_2F_5$	NH2	CH(CH ₃ )C=CCH ₃	ŏ		708070100	5_v11_1 +	ifluoromethyl 1 2	$r_{r}$
	$\tilde{C_2F_5}$	$NH_2$	CH2C CC2H5	0		dronumini	$-J - y_1 - + - u_1$	$(20 \text{ m})^{-1,2}$	,, ,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
	$C_2F_5$	NH ₂	$CH(CH_3)C \equiv CC_2H_5$	0		(10 -)	4h-1	$(0.5 \circ) = 1$	n.
	$C_2F_5$	NH ₂	$CH_2C \equiv CC_3H_7(n)$	0		(10 g), me	uriyi 10010(	= (0.5  g)  and sodiu	.m
	C2F5	NH2 NU-	$CH(CH_3)C = CC_3H_7(n)$	0	<i>(</i> <b>-</b>	were adde	and the	resultant mixture	W
	C2F5	NH ₂	CHOCoHs	ő	65	to 50° C. 1	or 3 hour	s. After completio	'n
	$C_2F_5$	NH ₂	$CH_2OC_3H_7(n)$	ŏ		the reaction	on mixture	e was poured into	١
	$C_2F_5$	NH ₂	$CH_2OC_3H_7(i)$	õ		with ethy	l acetate, a	and the organic la	łу
	$C_2F_5$	$NH_2$	$CH_2OC_4H_9(n)$	0		trated und	ler reduced	d pressure. The res	ic

 $|_{\mathbb{R}^2}$ Y (s) (i)  $CH_3$  $C_2H_5$  $C_3H_7(n)$  $C_3H_7(i)$  $C_4H_9(n)$  $C_4H_9(s)$  $C_4H_9(s)$ C4H9(i) H₂OC₂H₅ I₂F F3 CH2 o 1 0

(I)

 $\mathbb{R}^1$ 

) have optical isomers, the scope of the inven-

roduction of the comhown in the following

yl-6-fluoro-2 (3H)benzol-1,2,3,6-tetrahy-

- ) in dimethylformamide nd potassium carbonate resultant mixture was urs. After completion of re was poured into watals were collected by The residue was purified o give 1-[3-sec-butyl-6-
- n-5-yl]-3-metyl -4-tripyrimidine-2,6-dione
- MHz]: 0.90 (t, J=7 Hz, 2.3 (m, 2H), 3.45 (s, 3H), 3 (d, J=6 Hz, 1H), 7.32

-3-propargyl-2(3H)benyl-1,2,3,6-tetrahyin dimethylformamide l sodium hydride (0.4 g) ixture was heated at 40° npletion of the reaction, d into water, extracted anic layer was concentrated under reduced pressure. The residue was purified

by column chromatography to give 1-[6-fluoro-3-propargyl-2(3H) -benzoxazolon-5-yl]-3-methyl -4-trifluoromethyl-1,2,3,6-tetrahydropyrimidine-2,6-dione (0.3 g).

Example 3

To a solution of 1-[6-fluoro-3-sec-butyl-2(3H)-benzothiazolon-5-yl]-4-trifluoromethyl-1,2,3,6-tetrahydropyrimidine-2,6-dione (2.0 g) in dimethylformamide (10 g), sodium hydride (0.3 g) and 2,4-dinitrophenoxyamine (1.8 g) were added, and the resultant mixture was ¹⁰ heated at 40 to 60° C. for 3 hours. After completion of the reaction, the reaction mixture was poured into water, extracted with ethyl acetate and concentrated. The residue was purified by silica gel column chromatography to give 1-[6-fluoro-3-sec-butyl-2(3H)-benzothiazo-¹⁵ lon-5-yl]-3-amino-4-trifluoromethyl-1,2,3,6-tetrahydropyrimidine-2,6-dione (0.4 g).

In the same manner as above, the compounds (I) as shown in Table 2 were obtained.



Compound No.	R ¹	R ²	R ³	Y	Physical property
1	CF ₃	CH ₃	CH ₃	S	m.p. 102-103° C.
2	CF ₃	CH ₃	(s)C4H9	S	m.p. 84-86° C.
3	CF ₃	CH ₃	(i)C ₃ H ₇	S	resinous
4	CF ₃	CH ₃	$(n)C_3H_7$	S	m.p. 180–181° C.
5	CF ₃	CH ₃	CH ₂ CH=CH ₂	S	m.p. 61–62° C.
6	$CF_3$	$CH_3$	CH ₂ C≡CH	S	resinous
7	CF3	CH3	СН₃ І СНС≡СН	s	resinous
8	CF3	CH3	CH ₃ I CHCH=CH ₂	S	m.p. 146–147° C.
9	$CF_3$	CH3	сн=с=сн ₂	s	resinous
10	CF3	CH3	$CH_2C=CH_2$ I Cl	S	resinous
11	CF ₃	CH ₃	CH ₂ CH ₂ F	s	m.p. 179–180° C.
12	CF ₃	CHa	CH2OCH3	ŝ	m.p. 166–167° C.
13	C ₂ H ₅	CH ₃	(s)C ₄ H ₉	S	m.p. 157-159° C.
14	CF ₃	$NH_2$	(i)C ₃ H ₇	s	resinous
15	CF ₃	$NH_2$	(s)C4H9	s	resinous
16	CF ₃	CH ₃	CH ₂ C≡CH	0	m.p. 108–111° C.

The starting compound (II) may be produced according to the following scheme:





wherein  $R^4$  is a  $C_1$ - $C_6$  alkyl group and  $R^1$ ,  $R^3$  and Y are each as defined above.

The reaction at each step in the above scheme will be hereinafter explained in detail.

(1) Preparation of the compound (II) from the compound (III):

The compound (II) may be produced by reacting the 35 compound (III) with a compound of the formula:

$$R^{1}(NH_{2})C = CHCOOR^{5}$$
 (V)

wherein  $\mathbb{R}^5$  is a C₁-C₆ alkyl group and  $\mathbb{R}^1$  is as defined 40 above usually in the presence of a dehydrogenating agent in an inert solvent at a temperature of about 0° to 200° C. for a period of about 0.5 to 10 hours.

In general, the compound (V) and the dehydrogenating agent are used respectively in amounts of about 1 to

45 1.2 equivalents and of about 1 to 1.2 equivalents to one equivalent of the compound (III). As the dehydrogenating agent, there may be used an inorganic base (e.g. sodium carbonate, potassium carbonate, sodium hydride), an alkali metal alkoxide (e.g. sodium methoxide, 50 sodium ethoxide), etc.

Examples of the inert solvent are aliphatic hydrocarbons (e.g. hexane, heptane, ligroin, petroleum ether), aromatic hydrocarbons (e.g. benzene, toluene, xylene), ethers (e.g. diethyl ether, diisopropyl ether, dioxane, 55 tetrahydrofuran, diethylene glycol dimethyl ether), tertiary amines (e.g. pyridine, triethylamine, N,N-diethylaniline, tributylamine, N-methylmorpholine), acid amides (e.g. formamide, N,N-dimethylformamide, acetamide), sulfur compounds (e.g. dimethylsulfoxide, sul-60 phorane), etc. These may be used solely or in combination.

After completion of the reaction, the reaction mixture is subjected to ordinary post-treatment. For instance, the reaction mixture is poured into water and 65 extracted with an organic solvent, followed by concentration. If desired, any conventional purification procedure such as chromatography, distillation or recrystallization may be applied to the resulting product.

The compound (II) may be methylated or aminated without isolation to give the compound (I).

A typical embodiment for preparation of the compound (II) is illustratively shown in the following example.

Example 4

To a solution of 3-isopropyl-6-fluoro-5-methoxycarbonylamino -2(3H)-benzothiazolone (2.8 g) in N,Ndimethylformamide (10 g), sodium hydride (0.4 g) and ethyl 3-amino-4,4,4-trifluorocrotonate (0.9 g) were 10 added, and the resultant mixture was heated under reflux for 3 hours. After cooling, the reaction mixture was poured into water and extracted with ethyl acetate. The organic layer was concentrated, and the residue was purified by column chromatography to give 1-[3-iso-15 propyl-6-fluoro-2(3H) -benzothiazolon -5-yl]-4-trifluoromethyl-1,2,3,6-tetrahydropyrimidine -2,6-dione (0.8 g).

H-NMR  $\delta$  (ppm) [CDCl₃, 60 MHz]: 1.55 (6H, d, J=7 Hz), 4.4–5.5 (1H. m). 6.17 (1H, s), 7.0 (1H, d, J=6 Hz), 20 7.25 (1H, d, J=9 Hz), 9.0-10.1 (1H, m).

In the same manner as above, the compounds (II) as shown in Table 3 were obtained.



(2) Preparation of the compound (III) from the compound (IV):

The compound (III) may be produced by reacting the compound (IV) with a compound of the formula:

$$\bigcup_{\substack{\parallel \\ ClCOR^5}}^{O} (VI)$$

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wherein  $\mathbb{R}^5$  is a  $\mathbb{C}_1$ - $\mathbb{C}_6$  alkyl group in the existence of a dehydrohalogenating agent in the presence or absence of an inert solvent at a temperature of about 0° to 150° C. for a period of about 0.5 to 10 hours.

Normally, the compound (VI) and the dehydrohalogenating agent are used respectively in amounts of about to 1.5 equivalents and of about 1 to 1.5 equivalents to one equivalent of the compound (IV). As the dehydrohalogenating agent, there may be used an or- 55 ganic base (e.g. pyridine, triethylamine, N,N-diethylaniline), an inorganic base (e.g. sodium hydroxide, potassium hydroxide, sodium carbonate, potassium carbonate, sodium hydride), an alkali metal alkoxide (e.g. sodium methoxide, sodium ethoxide), etc.

Examples of the inert solvent are aromatic hydrocarbons (e.g. benzene, toluene, xylene), halogenated hydrocarbons (e.g. chloroform, carbon tetrachloride), ethers (e.g. diethyl ether, dioxane), etc.

After completion of the reaction, the reaction mix- 65 ture is subjected to ordinary post-treatment. For instance, the reaction mixture is poured into water and extracted with an organic solvent, followed by concen-

tration. If desired, any conventional purification procedure such as chromatography, distillation or recrystallization may be applied to the resulting product.

A typical embodiment for production of the compound (III) is illustratively shown in the following Example.

Example 5

A mixture of 5-amino-6-fluoro-3-isopropyl-2(3H)benzothiazolone (2.1 g), N,N-diethyianiline (1.5 g) and methyl chloroformate (1.0 g) was dissolved in 1,2dichloroethane (10 g), and the resultant mixture was heated under reflux for 3 hours. After cooling, the reaction mixture was washed with water, and the organic layer was concentrated. The residue was washed with methanol to give 6-fluoro-5-methoxycarbonylamino -3-isopropyl-2(3H)-benzothiazolone (2.1 g).

In the same manner as above, the compounds (III) as shown in Table 4 were obtained.



The compound (IV) can be produced by the method as disclosed in U.S. Pat. No. 4,640,707 or U.S. Pat. No. 15 4,720,297.

For the practical usage of the compound (I), it is usually formulated with conventional solid or liquid carriers or diluents as well as surface active agents or auxiliary agents into conventional preparation forms such as emulsifiable concentrates, wettable powders, suspensions, water dispersible granules and granules. The content of the compound (I) as the active ingredient in such preparation forms is normally within a range of about 0.02 to 80% by weight, preferably of about 0.05 to 70% by weight. Examples of the solid carrier or diluent are fine powders or granules of kaolin clay, attapulgite clay, bentonite, terra alba, pyrophyllite, talc, diatomaceous earth, calcite, walnut powders, urea, ammonium sulfate and synthetic hydrous silicate, etc. As the liquid carrier or diluent, there may be exemplified aromatic hydrocarbons (e.g. xylene, methylnaphthalene), alcohols (e.g. isopropanol, ethylene glycol, cellosolve), ketones (e.g. acetone, cyclohexanone, isophorone), soybean oil, cotton seed oil, dimethylsulfoxide, N,N-dimethylformamide, acetonitrile, water, etc.

The surface active agent used for emulsification, dispersion or spreading may be of any type, for instance, either anionic or non-ionic. Example of the surface active agent include alkylsulfates, alkylarylsulfonates, dialkylsulfosuccinates, phosphates of polyoxyethylenealkylaryI ethers, polyoxyethylene alkyl ethers, polyoxyethylene alkylaryl ethers, polyoxyethylene 5 polyoxypropylene block copolymer, sorbitan fatty acid esters, polyoxyethylene sorbitan fatty acid esters, etc. Examples of the auxiliary agents include ligninsulfonates, sodium alginate, polyvinyl alcohol, gum arabic, CMC (carboxymethyl celluloses, PAP (isopropyl acid 10 phosphate), etc.

Practical embodiments of the herbicidal composition according to the present invention are illustratively shown in the following examples wherein parts are by weight. The number of the active ingredient corre- 15 sponds to the one in Table 2.

Formulation Example 1

Fifty parts of any one of Compound Nos. 1, 2, 4. 5, 8, 11 to 13 or 16, 3 parts of calcium ligninsuifonate, 2 parts of sodium laurylsulfate and 45 parts of synthetic hy- 20 drous silicate are well mixed while being powdered to obtain a wettable powder.

Formulation Example 2

Five parts of any one of Compound Nos. 1 to 16, 14 parts of polyoxyethylenestyrylphenyl ether, 6 parts of 25 calcium dodecylbenzenesulfonate, 25 parts of xylene and 50 parts of cyclohexanone are well mixed to obtain an emulsifiable concentrate.

Formulation Example 3

Two parts of any one of Compound Nos. 1 to 16, 1 30 part of synthetic hydrous silicate, 2 parts of calcium ligninsulfonate,. 30 parts of bentonite and 65 parts of kaolin clay are well mixed while being powdered. The mixture is then kneaded with water, granulated and dried to obtain granules.

Formulation Example 4

Twenty-five parts of any one of Compound Nos. 1, 2, 4, 5, 8, 11 to 13 or 16 are mixed with 3 parts of polyoxyethylene sorbitan monooleate, 3 parts of carboxymethyl cellulose and 69 parts of water and pulverized until the 40 particle size of the mixture becomes less than 5 microns to obtain a suspension.

Formulation Example 5

0.05 Part of any one of Compound Nos. 1 to 16, 1 part of synthetic hydrous silicate, 2 parts of calcium lignin- 45 sulfonate, 30 parts of bentonite and 66.95 parts of kaolin clay are well mixed while being powdered. The mixture is then kneaded with water, granulated and dried to obtain granules.

The compound (I) thus formulated in any suitable 50 preparation form is useful for pre-emergence or postemergence control of undesired weeds by soil or foliar treatment as well as flood fallowing treatment. These treatments include application to the soil surface prior to or after planting, incorporation into the soil prior to 55 planting or transplanting, etc. The foliar treatment may be effected by spraying the herbicidal composition containing the compound (I) over the top of plants. It may also be applied directly to the weeds if care is taken to keep the chemical off the crop foliage. 60

The compound (I) may be used together with any other herbicide to improve its activity as a herbicide, and in some cases, a synergistic effect can be expected. Further, it may be applied in combination with an insecticide, an acaricide, a nematocide, a fungicide, a plant 65 cm) were filled with upland field soil, and the seeds of growth regulator, a fertilizer, a soil improver, etc. It is also useful as a herbicide to be employed for orchards, pasture lands, lawns, forests, non-agricultural fields, etc.

The dosage of the compound (I) may vary depending on the prevailing weather conditions, the formulation used, the prevailing season, the mode of application, the soil involved, the crop and weed species, etc. Generally, however, the dosage is from about 0.01 to 80 grams, preferably from about 0.02 to 40 grams, of the active ingredient per are. The herbicidal composition of the invention formulated in the form of an emulsifiable concentrate, a wettable powder, a water-dispersible granule or a suspension may ordinarily be employed by diluting it with water at a volume of about 1 to 10 liters per are, if necessary with addition of an auxiliary agent such as a spreading agent. Examples of the spreading agent include, in addition to the surface active agents as noted above, polyoxyethylene resin acid (ester), ligninsulfonate, abietylenic acid salt, dinaphthylmethanedisulfonate, paraffin, etc. The composition formulated in the form of granules may be normally applied as such without dilution.

The biological data of the compound (I) as herbicides will be illustratively shown in the following Examples wherein the phytotoxicity to crop plants and the herbicidal activity on weeds were observed visually as to the degree of germination as well as the growth inhibition and rated with an index 0, 1, 2, 3, 4, or 5, the numeral "0" indicating no material difference as seen in comparison with the untreated plants and the numeral "5" indicating the complete inhibition or death of the test plants.

The compounds as shown in Table 5 were used for comparison.

TABLE 5



#### Test Example 1

Cylindrical plastic pots (diameter, 10 cm; height, 10 Japanese millet, oats, tall morningglory and velvetleaf were sowed therein and covered with soil. A designated amount of the test compound formulated in an emulsifi-

able concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed onto the soil surface by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were grown in a greenhouse for 20 days, and the herbicidal activity 5 was examined. The results are shown in Table 6.

TABLE 6

			Herbi	cidal activity		
Compound No.	Dosage (g/are)	Japanese millet	Oats	Tall morning- glory	Velvet- leaf	10
1	5	5	5	5	5	
	1.25	5	5	5	5	
2	5	5	5	5	5	
	1.25	5	5	5	5	15
3	5	5	5	5	5	
	1.25	5	5	5	5	
4	5	5	5	5	5	
	1.25	5	5	5	5	
5	5	5	5	5	5	
	1.25	5	5	5	5	20
6	5	5	5	5	5	20
	1.25	5	5	5	5	
7	5	5	5	5	5	
	1.25	5	5	5	5	
8	5	5	5	5	5	
	1.25	5	5	5	5	25
8	5	5	5	5	5	20
	1.25	5	5	5	5	
9	5	5	5	5	5	
	1.25	5	5	5	5	
10	5	5	5	5	5	
	1.25	5	5	5	5	• •
11	5	5	5	5	5	30
	1.25	5	5	5	5	
12	5	5	5	5	5	
	1.25	5	5	5	5	
13	5	5	5	5	5	
	1.25	5	5	5	5	
14	5	5	5	5	5	35
	1.25	5	5	5	5	
15	5	5	5	5	5	
	1.25	5	5	5	5	
16	5	5	5	5	5	
	1.25	5	5	5	5	
Α	1.25	1	0	1	3	40
в	2.5	0	0	0	0	
	1.25	0	0	0	0	
С	1.25	0	0	0	0	1

Test Example 2

Cylindrical plastic pots (diameter, 10 cm; height, 10 ⁴³ cm) were filled with upland field soil, and the seeds of Japanese millet, tall morningglory, radish, velvetleaf and oats were sowed therein and cultivated in a greenhouse for 10 days. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 was diluted with water containing a spreading agent, and the dilution was sprayed over the foliage of the test plant by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were further grown in the greenhouse for 20 days, and the herbicidal activity was examined. The results are shown in Table 7.

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	-		Herbicidal activity							
Com- pound No.	Dosage (g/are)	Japanese millet	Tall morning- glory	Radish	Velvet- leaf	Oats				
1	10	5	5	5	5	5	-			
	1.25	5	5	5	5	5	65			
2	10	5	5	5	5	5	05			
	1.25	5	5	5	5	5				
3	10	5	5	5	5	5				
	1.25	5	5	5	5	5				

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			Herb	icidal acti	vity						
Com- pound No.	Dosage (g/are)	Japanese millet	Tall morning- glory	Radish	Velvet- leaf	Oats					
4	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
5	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
6	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
7	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
8	10	5	5	5	5	5					
	1.25	5	. 5	5	5	5					
9	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
10	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
11	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
12	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
13	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
14	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
15	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
16	10	5	5	5	5	5					
	1.25	5	5	5	5	5					
А	1.25	3	3	5	5	1					
В	1.25	0	0	0	0	0					
С	10	0	2	1	3	0					
	1.25	0	1	0	1	0					

Test Example 3

Cylindrical plastic pots (diameter, 8 cm.; height, 12 cm) were filled with paddy field soil, and the seeds of barnyardgrass (Echinochloa oryzicola), broad-leaved weeds (i.e. common falsepimpernel, indian toothcup, waterwort) were sowed in 1 to 2 cm depth. Water was poured therein to make a flooded condition, and rice seedlings of 3-leaf stage were transplanted therein, and the test plants were grown in a greenhouse. Six days (at that time weeds began to germinate) thereafter, a designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 and diluted with water (5 ml) was applied to the pots by 45 perfusion. The test plants were grown for an additional 20 days in the greenhouse, and the herbicidal activity and phytotoxicity were examinated. The results are shown in Table 8.

TABLE 8

		Phyto- toxicity	Herbic	idal activity
Compound No.	Dosage (g∕are)	Rice plant	Barnyard- grass	Broad-leaved weed
2	0.16	1	5	5
3	0.16	1	5	5
4	0.16	1	5	5
5	0.16	1	5	5
6	0 16	1	5	5
7	0.16	1	5	5
8	0.16	1	5	5
9	0.16	1	5	5
10	0.16	1	5	5
11	0.16	1	5	5
12	0.16	1	5	5
13	0.16	2	5	5
16	0.16	2	4	5
Α	0.16	0	2	2
В	0.16	0	2	2
С	0.16	0	0	0

Vats  $(33 \text{ cm} \times 23 \text{ cm} \times 11 \text{ cm})$  were filled with upland

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Test Example 4

activity and phytotoxicity were examined. The results are shown in Table 10.

				Herbicidal activity				
Compound	Dosage	Phytot	oxicity	Pale	Common	Persian	Field	
No.	(g/a)	Wheat	Barley	smartweed	chickweed	speedwell	pansy	
1	0.63	0	1	5		4	5	
2	0.63	0	0	5	5	5	5	
	0.32	0	0	5	5	5	5	
3	0.63	1	1	5	4	5	5	
4	0.63	—	1	5	5	5	5	
5	0.63	1	1	5	5	5	5	
6	0.63	1	-	5	5	5	5	
	0.32	0	1	5	5	5	5	
7	0.63			5	5	5	5	
	0.32	0	0	5	4	5	5	
8	0.63	0	0	5	4	5	_	
9	0.63	0	0	5	5	5	_	
13	0.63	0	0	5	4	4	5	
14	0.63			5	5	5	5	
	0.32	1	1	5	5	5	5	
15	0.63		1	5	5	5	5	
	0.32	1	1	5	5	5	5	
С	0.63	0	0	0	0	0	0	

field soil, and the seeds of soybean, corn, rice plant, velvetleaf, common cocklebur, tall morningglory, black ²⁵ nightshade, redroot pigweed and green foxtail were sowed therein in 1 to 2 cm depth. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed onto the soil ³⁰ surface by means of a small hand sprayer at a spray volume of 10 liter per are. The test plants were groton in a greenhouse for 20 days, and the herbicidal activity and phytotoxicity were examined. The results are shown in Table 9. ³⁵ Test Example 6

Vats  $(33 \text{ cm} \times 23 \text{ cm} \times 11 \text{ cm})$  were filled with upland field soil, and the seeds of corn, common cocklebur, velvetleaf, tall morningglory, black nightshade, barnyardgrass and green foxtail were sowed therein and cultivated for 18 days in a greenhouse. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed over the foliage of the test plants by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were further grown in the greenhouse for 20

TABLE 9

					Herbicidal activity						
		Ph	ytotoxi	city		Common	Tall	Black			
Compound No.	Dosage (g/a)	Soy- bean	Corn	Rice plant	Velvet leaf	cock- lebur	morning- glory	night- shade	Redroot pigweed	Green foxtail	
2	1.25	1	0	0	5	5	5	5	5	5	
5	1.25	1	0	0	5		5	5	5	5	
13	1.25	1	0	_	5	_	4	5	5	5	
16	1.25	1	0	0	5	_	5	5	5		
С	1.25	0	0	0	0	0	0	0	0	0	

Test Example 5

Vats  $(33 \text{ cm} \times 23 \text{ cm} \times 11 \text{ cm})$  were filled with upland field soil, and the seeds of wheat, barley, pale smartweed, common chickweed, persian speedwell and field pansy were sowed therein in 1 to 2 cm depth. A designated amount of test compound formulated in an emul-

days, and the herbicidal activity and phytotoxicity were examined. At the time of the application, the test plants were generally at the 1 to 4 leaf stage and in 2 to 12 cm height, although the growing stage of the test plants varied depending on their species. The results are shown in Table 11.

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			Herbicidal activity					
Compound No.	Dosage (g/a)	Phyto- toxicity Corn	Common cock- lebur	Velvet- leaf	Tall morning- glory	Black night- shade	Barn- yard- grass	Green fox- tail
2	0.08	1	5	5	5	5	4	4
7	0.08	1	5	5	5	5	4	4
6	0.08	1	5	5	5	5	5	5
С	0.08	0	0	0	0	0	0	0

sifiable concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed onto 65 the soil surface by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were grown in a greenhouse for 27 days, and the herbicidal

#### Test Example 7

Vats  $(33 \text{ cm} \times 23 \text{ cm} \times 11 \text{ cm})$  were filled with upland field soil, and the seeds of wheat, barley, pale smartweed, catchweed bedstraw, common chickweed, persean speedwell and field pansy were sowed therein and

cultivated for 25 days in a greenhouse. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed over the foliage of the test plants by means of a small hand 5 sprayer at a spray volume of 10 liters per are. The test plants were further grown in the greenhouse for 27 days, and the herbicidal activity and phytotoxicity were examined. At the time of the application, the test plants were generally at the 1 to 4 leaf stage and in 2 to 12 cm 10 height, although the growing stage of the test plants varied depending on their species. The results are shown in Table 12.

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of the treatment, the depth of water in the pots was kept at 4 cm and following two days, water was let leak a volume corresponding to a 3 cm depth per day.

	TABLE 13											
		Phyto- toxicity	Herbic	idal activity								
Compound	Dosage	Rice	Broad-	Barnyard-								
NT -	(- ()		leaved									
INO	(g/are)	plant	weed	grass								
4	0.04	1	5	5								
5	0.04	1	5	4								
6	0.04	1	5	5								
7	0.04	0	5	5								

Herbicidal activity											
Compound	Dosage	Phytot	oxicity	Pale smart-	Catch weed	Common chick	Persian speed-	Field			
No.	(g/a)	Wheat	Barley	weed	bedstraw	weed	well	pansy			
1	0.08	1	1	5	5	5	5	5			
2	0.08	1	1	5	5	5	5	5			
3	0.08	1	0	5		4	4	4			
4	0.08	0	1	5	5	4	5	5			
5	0.08	0	1	5	4	4	5	5			
6	0.08	1	1	5	4	5	5	5			
7	0.08	1	1	4	4	4	5	5			
8	0.08	0	0	5		4	5	5			
9	0.08	1	1	4	4	_	5	5			
10	0.08	0	0	5		4	5	5			
14	0.08	1	1	5	_		5	5			
15	0.08	1	1	5			5	5			
16	0.08	1		5	4	5	5	5			
Α	0.08	0	0	0	0	0	1	1			
в	0.08	0	0	0	0	0	0	0			
С	0.08	0	0	0	0	0	0	0			

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#### Test Example 8

Wagner's pots (1/5000 are) were filled with paddy ³⁵ field soil, and the seeds of broad-leaved weed (e.g. common falsepimpernel, indian toothcup, waterwort) and barnyardgrass were sowed in 1 to 2 cm depth. Water was poured therein to make a flooded condition, and 40 rice seedlings of 2-leaf stage were transplanted therein, and the test plants were grown in a greenhouse. Four days thereafter, a designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 and diluted with water (10 ml) 45 was applied to the pots by perfusion. The test plants were grown for an additional 20 days in the greenhouse, and the herbicidal activity and phytotoxicity were examined. The results are shown in Table 13. At the time

#### Test Example 9

Vats  $(33 \text{ cm} \times 23 \text{ cm} \times 11 \text{ cm})$  were filled with upland field soil, and the seeds of corn, rice plant, velvetleaf, tall morningglory, black nightshade, redroot pigweed and green foxtail were sowed therein in 1 to 2 cm depth. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formulation Example 2 was diluted with water, and the dilution was sprayed onto the soil surface by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were grown in a greenhouse for 20 days, and the herbicidal activity and phytotoxicity were examined. The results are shown in Table 14.

TABLE 14

				Herbicidal activity				
		Phyto	toxicity		Tall	Black		
Compound No.	Dosage (g/a)	Corn	Rice plant	Velvet- leaf	morning- glory	night- shade	Redroot pigweed	Green foxtail
1	1.25	0	1	5	5	5	5	4
2	1.25	0	0	5	5	5	5	5
	0.32	0	0	5	4	5	5	4
3	1.25	1		5	5	5	5	5
	0.32	0	0	5	4	5	5	5
4	1.25	1	—	5		5	5	5
5	1.25	0	0	5	5	5	5	5
6	1.25	0	0	5	5	5	5	5
	0.32	0	0	5	4	5	5	4
7	1.25	1	0	5	5	5	5	5
	0.32	0	0	5	4	5	5	4
8	1.25	0	0	5	4	5	5	5
10	1.25	1	1	5	5	5	5	5
14	1.25	1		5	4	5	5	5
	0.32	1		5	4	5	5	
15	1.25	1	—	5	5	5	5	5
	0.32	0	1	5	5	5	5	4.

TABLE 14-continued

					Herbicidal activity				
		Phyto	Phytotoxicity		Tall	Black			
Compound No.	Dosage (g/a)	Corn	Rice plant	Velvet- leaf	morning- glory	night- shade	Redroot pigweed	Green foxtail	
С	1.25	0	0	0	0	0	0	0	

Test Example 10

field soil, and the seeds of corn, common cocklebur, velvetleaf, tall morningglory and black nightshade were sowed therein and cultivated for 18 days in a greenhouse. A designated amount of the test compound formulated in an emulsifiable concentrate as in Formula- 15 tion Example 2 was diluted with water, and the dilution was sprayed over the foliage of the test plants by means of a small hand sprayer at a spray volume of 10 liters per are. The test plants were further grown in the greentoxicity were examined. At the time of the application, the test plants were generally at the 1 to 4 leaf stage and in 2 to 12 cm height, although the growing stage of the test plants varied depending on their species. The results are shown in Table 15. 25

wherein R¹ is a trifluoromethyl group or a pentafluoro-Vats (33 cm $\times$ 23 cm $\times$ 11 cm) were filled with upland 10 ethyl group, R² is an amino group, R³ is a C₁-C₇ alkyl group, a C₃-C₇ alkenyl group, a C₃-C₇ alkynyl group, a halo  $(C_1-C_6)$  alkyl group, a halo $(C_3-C_6)$  alkenyl group or a  $C_1$ - $C_4$  alkoxy( $C_1$ - $C_3$ ) alkyl group and Y is an oxygen atom or a sulfur atom.

> 2. The compound according to claim 1, wherein Y is a sulfur atom.

> 3. The compound according to claim 1, wherein  $\mathbb{R}^3$  is a  $C_1$ – $C_7$  alkyl group.

4. A herbicidal composition which comprises as an house for 20 days, and the herbicidal activity and phyto- 20 active ingredient a herbicidally effective amount of the compound according to claim 1, and an inert carrier or diluent.

> 5. The compound according to claim 1, wherein  $\mathbf{R}^1$  is  $OF_3$ ,  $R^2$  is  $-NH_2$ ,  $R^3$  is  $-(i)C_3H_7$  and Y is sulfur.

> 6. The compound according to claim 1, wherein  $\mathbb{R}^1$  is

				Herbicidal	activity	
Compound No.	Dosage (g/a)	Phytotoxicity Corn	Common cocklebur	Velvet- leaf	Tall morning- glory	Black night- shade
1	0.08	1	5	5	5	5
2	0.08	1	5	5	5	5
	0.02	1	5	5	4	5
3	0.08	1	5	5	5	5
4	0.08	1	5	5	5	5
5	0.08	1	5	5	5	5
6	0.08	1	5	5	5	5
	0.02	1	4	5	5	5
7	0.08	1	5	5	5	5
	0.02	1	4	5	5	5
8	0.08	1	_	5	4	5
9	0.08	1	5	5	5	5
10	0.08	1	5	5	5	5
13	0.08	1	5	5	5	5
14	0.08	1	5	5	5	5
	0.02	0	4	5	4	5
15	0.08	1	5	5	5	5
	0.02	0	4	5	_	5
С	0.08	0	0	0	0	0

TABLE 15

What is claimed is:

1. (Amended) A compound of the formula:



 $-OF_3$ ,  $R^2$  is  $-NH_2$ ,  $R^3$  is  $-(s)C_4H_9$  and Y is sulfur. 7. The compound according to claim 1, wherein Y is ⁵⁰ an oxygen atom.

8. The compound according to claim 3, wherein Y is a sulfur atom.

9. The compound according to claim 3, wherein Y is an oxygen atom.

55 10. The compound according to claim 1, wherein Y is a sulfur atom and  $R^3$  is a C₁-C₇ alkyl group or a C₃-C₇ alkynyl group.

11. The compound according to claim 1, wherein Y is a oxygen atom and  $\mathbb{R}^3$  is a  $\mathbb{C}_1$ - $\mathbb{C}_7$  alkyl group or a 60 C₃-C₇ alkynyl group.